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KINETICS OF SUBBITUMINOUS COAL DRYING*

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ABSTRACT

This paper explores both the effects of stress upon water removal from subbituminous coal and the consequences of drying upon underground coal conversion. Laboratory tests studying compressive stress effects on moisture transfer in the hygroscopic region are reported. Measurements of CO_2 permeability are also reported as a function of compressive stress and fluid saturation levels. Results indicate that molecular transport phenomena are unaffected by compressive stress levels while viscous transport is markedly influenced by stress. This flow decrease results from alteration of the size of larger flow channels which are necessary to support viscous fluid transport.

Introduction

There is considerable evidence to suggest that saturated coal, in the natural, stressed underground condition, has low permeability. (1) The movement of fluids through a coal mass occurs principally through fracture systems in that particular section. Because flow dynamics through fracture distributions follows the mathematics of log-normal distributions, the greatest majority of mass transfer occurs through a single group of fractures. Underground coal conversion involves a complex thermal transfer process in a coal mass. The rates and mechanisms of this transfer are closely coupled to the kinetics of moisture removal from coal. Consequently, it is important to learn about moisture removal dynamics.

Subbituminous coals, the coals of interest for Western underground coal conversion, are hygroscopic due to the polar, oxygenated groups (-OH, -COOH, etc.) incorporated within the carbonaceous structure. Typically these coals contain significant quantities of water and seams show appreciable water transmittivity, e.q., they can be productive aquifers. (2) Water present in these coals is in more than one form. (3) Underground water is contained in cracks, fissures and capillaries. Such water may be held by dispersion forces, held in close proximity to the surface of pore structures, or such water may be contained in larger flow channels. (Obviously, other water is contained as part of the coaly material effectively removed from any flow channels. We do not consider that material here). In this discussion, water in coal will be considered present only in two forms: a.) water in cracks and capillaries will be termed ''excess water'' and physically adsorbed water will be termed ''adsorbed water''. The physical distinction between these types may be clear although the boundary between them is not. One possibility for distinction is on the physical geometries of the flow channels. Sometimes openings smaller than 10^{-5} cm, have been used for this distinction, but openings under 10^{-7} cm are probably more reasonable. (4,5) This size distinction offers a convenient definition to differentiate between excess and adsorbed water.

The interactions between stress, flow channels and fluid flow in underground coal processing are complicated. Naturally occurring coals are stressed by both pore pressure and lithostatic pressure forces. Coal is an elastic material (6) and it is possible that increasing stress will deform flow channels. Depending upon the depth of the seam and other factors increasing stress could be predicted to either increase or decrease mass transfer rates. At more shallow depths, stress increases could increase porosity due to lifting of the overburden layers. At deeper locations, stress increases may well be contained in the coal seam due to higher lithostatic forces. In that case, plastic deformations should lead to a rearrangement in porosity closing off particular flow channels and enlarging others, i.e., building a fissure system. Water concentrations in coal,

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calculated on a weight/weight basis, are time dependent and highly heterogeneous. There is some correlation that drier coals are located in regions with scant annual surface precipitation (compare San Juan Basin with Powder River Basin), but that correlation is probably simplistic. Point moisture concentrations represent a steady-state kinetic process, i.e., the difference between water influx rates and water removal rates. The fact that removal rates may be fairly constant and influx rates less so suggests that seam moisture will vary over long times.

Moisture removal from subbituminous coals leads to a sharply increased porosity. (6) This porosity results from a stress-cracking process. Initially, moisture removal appears reversible. It is possible that this first porosity change simply reflects excess water variation. Due to the elastic nature of natural coal, dewatering may result in subsidence as the lithostatic forces close fissures in the dewatered regions. However, dried coal exhibits brittle behavior and considerable strength. (6) Consequently, during the drying process, coal properties undergo an elastic-brittle transition, and lose the ability to plastically deform.

During UCC, the removal of water from a coal seam takes place in two fairly distinct steps: First, then is drainage of excess water by pumping, followed by adsorbed moisture removal from the bulk coal through the free surfaces of fissures, via forced convection of air or hot gases (6).

1. Coal Seam Drainage. Here liquid water residing in the supporting network of fissures is removed by viscous flow. It must be recognized that not all excess water is readily removed. Body forces are not sufficient to completely overcome capillary forces (in a capillary-porous body such as cracked coal). Hence only some fraction of the liquid can be removed by direct pumping. Also, since the overburden is saturated, there is potential for replacement (''water influx''). It should be noted that excess water removal causes local changes in stress and permeability which are large in magnitude and which can alter water flow patterns.

2. Adsorbed Moisture Removal. The removal of adsorbed water is viewed as a two-step process. Via forced convection of air or hot gases, removal begins with desorption of the water molecules from the coal's inner porous structure. This is followed by molecular diffusion of H_2O, through pores of molecular dimension until a fissure is reached which is large enough to support viscous flow of the fluid into the main fissure network. (Obviously, since blind pores and cracks are likely to be present, a series of desorption-diffusion-adsorption events must take place giving rise to the appearance of surface flow).

The thermodynamic driving potential for the migration of this adsorbed moisture to the free surface is concentration. Once the surface concentration is lowered by passage of air (<100% RH) or a hot gas, random movement of adsorbed moisture begins to flow to that surface. This concentration gradient is accompanied by a stress gradient due to moisture expansion of the coal material. Since coal has low tensile strength, this concentration gradient is adequate to induce cracking, and hence, extends the fissure network, limited by local lithostatic or hydrostatic stresses.

The research described here is aimed at the eventual understanding of *in situ* coal drying processes and concommitent permeability and stress field changes. In particular, the hygroscopic region of moisture saturation (adsorbed moisture) has been studied. The effects of stress and drying temperature have been investigated by two methods: a) determination of the effective diffusion coeffecient of water vapor in coal under a transient compressive stress and b) determination of the effective permeability to carbon dioxide under moist and dry condition and varying compressive stress. Coals used were subbituminous C from the States of Washington and New Mexico, respectively.

Experimental Investigations

Changes of Permeability with Compressive Stress and Moisture Content

Measurements underway study the influence of stress on mass transfer in coal. These studies use carbon dioxide gas as the working fluid and make measurements in rapid enough times so that water contents are essentially constant. Apparatus for these measurements is shown in Figure 1. Coal samples, right cylinders cored from a freshly removed Fruitland sample, are confined within an elastic (neoprene) sleeve. This sleeve with sample is inserted into a tightly fitting steel cylinder. Geometries are so arranged that the neoprene confining sleeve is slightly longer than the steel cylinder. Coal samples are typically cylinders 5 cm in diameter and 4 cm long. The assembly containing the coal sample is positioned between two platens of a hydraulic system. Transverse force is transferred through the neoprene elastic sleeve into radial confining stress on the coal sample. Sample gas is transferred from one "ballast volume" to the other. Pressures are measured in a dual-differential mode so that gas volumes both going into and coming from the sample can be independently measured.

Data were taken using coal with proximite analysis typical for the San Juan Basin

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(Fruitland) (7). Moisture content, as received, was approximately 9.5%

Results suggest that stress markedly changes gas permeability. Coal samples, measured with no applied radial stress show CO_2 permeability in the region of 10 to 100 md. Examination of these samples showed visible micro fractures, aperatures in the size range of 0.1 mm. Placing confining stress on these samples closed aperatures to a far smaller size.

Data showing permeability changes induced by stress loading are shown in Figure 2. Initial flow apparently is through microfractures and other "fissure" flow channels. Stress loading changes permeability from approximately 10 md, to, at equivalent stress loading to 500 feet of overburden, 0.02 md. This factor of $10^{2.6}$ decrease is partially inelastic. Curve B, Figure 2, shows similar data following measurements given in Curve A. The coal, following removal of stress, does not return to the initial flow configuration. Rather the second series of measurements is replicated. This suggests that confining stress is sufficient to seal, at least temporarily, coal microfractures. Since naturally occurring coals are continuously in compression due to the lithostatic stress, few microfractures should exist.

These data were obtained on wet (as received) samples. Data were also obtained following moisture removal. Here CO_2 gas flow is particularly effective in moisture removal. Water drainage is followed by quantitative measurement of water recovered from a cold trap incorporated in the apparatus. Data obtained on dried samples are shown in Figure 3. Data in Figure 3, upper curve, show results obtained after approximately 60% of the total moisture that could be removed by drying at 110°C was recovered by flowing CO_2 through the sample at 25°C. The gas permeability values were far higher, by a factor of five, at zero applied stress. Again, with confining stress the permeability decreased; however, the effect was less significant. Dried coal, under the same pressure interval used to stress wet coal, showed a permeability decrease of $10^{1.6}$. It appears that the elastic nature of the material is reduced well before the coal is completely dry.

Changes in Subbituminous Coal Porosity with Moisture Content

It is well known that subbituminous coal shrinks with drying. (8) This results from tensile cracks induced by localized dimensional changes. Removal of excess water from fissure systems in samples under stress does not generate appreciable porosity; this is apparent from the previous section. Adsorbed water removal is required for significant porosity generation.

It is necessary to know much more about thermally-induced porosity generation in these materials. The boundary between wet and dry coal is a boundary between low porosity and high porosity material. Heat transfer mechanisms are appreciably different on two sides of this boundary due to the sharp porosity changes. Data were obtained on a section of Fruitland Seam to determine connected porosity. A right cylindrical section, approximately 10 cm in diameter and 6 cm thick was inserted in a 10.2 cm I.D. steel pipe and cemented in that pipe with epoxy (Epon) cement. The coal-steel was then cut to make a sample of 6-cm thickness. This was inserted into a Boyle's Law porosity instrument. Initial measurements were made on the wet sample. Data showed a porosity of 1.8%. The sample was then removed from the instrument and inserted into an oven at 90°C and moisture was partially removed. Porosity, at 25°C was then measured again. Moisture loss was determined gravimetrically. This procedure was repeated until 7.7% of the total sample weight was removed. (This coal sample contained, initially, 9.6% moisture).

These data are shown in Figure 4. The porosity generation rate is initially about twice the rate of water removal, i.e., the removal of 1% of the total water (that removed at 100°C) results in a 2% porosity increase. This changes in the region of 50% total water removed to, finally, result in a dry porosity that is similar in magnitude to the initial water content. For instance, data in Figure 4 show that this coal sample showed a final porosity of approximately 10%.

These data were obtained at ambient stress conditions. They, therefore, must be taken as preliminary because applied stress may well change the porosity generation rates. However they do show the transition state that must exist at the wet-dry coal boundary. (9)

At the "wet" side, little connected porosity is available to serve as mass transfer channels. Drying the coal then alters this situation forming a zone of highly porous, and, of course, permeable coal. This porosity generation and its influence on mass transfer is a key element in the control of underground coal conversion.

Variation of Effective Diffusion Coefficient of Water in Coal with Transient Compressive Stress and Temperature

The preceding results both show the influence of stress on the alteration of mass transfer channels in coal and describe the motion of excess water through coal seams. The evidence is clear that such channels are readily altered by the external stress state until they are locked into

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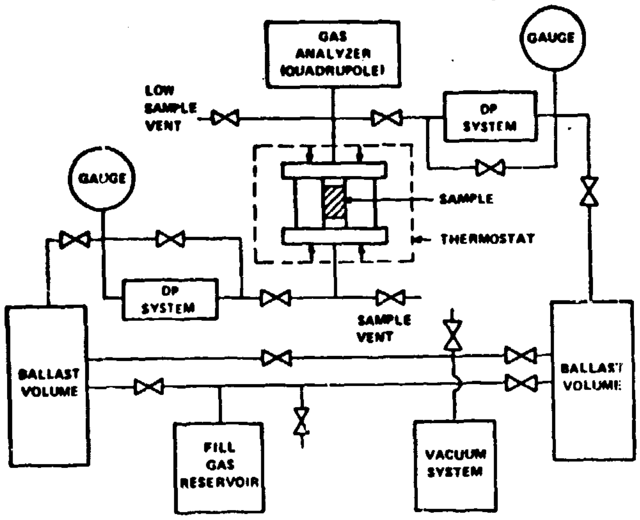

Figure 1: Sche..atic of apparatus used to measure stress effects on permeability. Dual differential pressure gauges measure pressure differences between sample and "ballast" pressures so that gas fluxes in and out of sample can be determined.

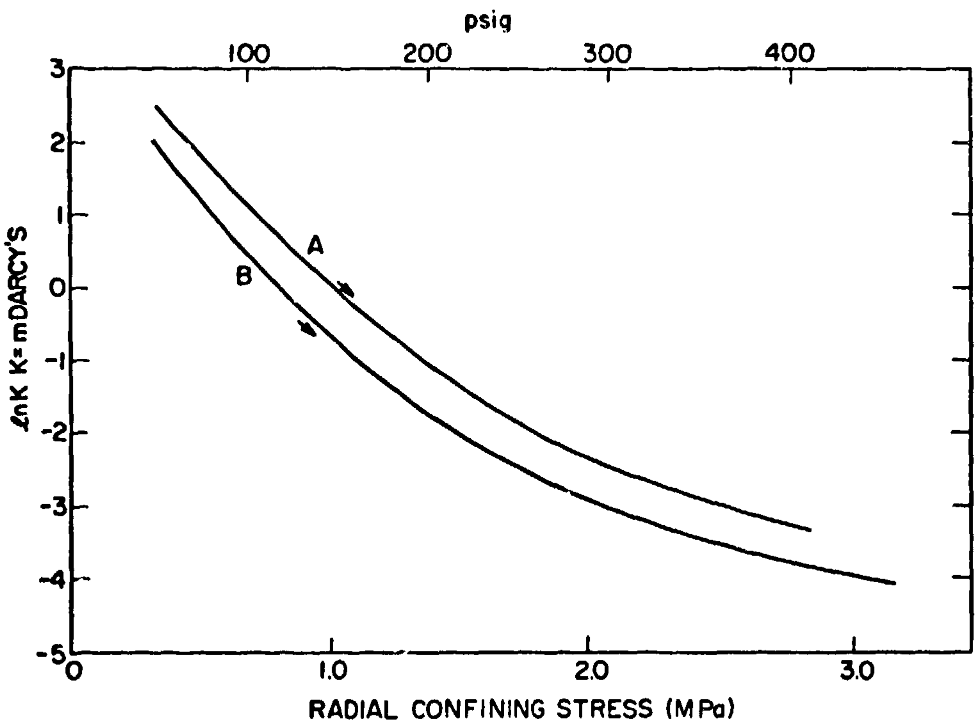

Figure 2: Variation of CO_2 permeability with confining stress on Fruitland Coal. Sample removed from Sage Pit, Fruitland Seam (Western Coal Company, Farmington, NM) Sample oriented so that flow is parallel to bedding. Moisture content, 9.5%. Curve A shows initial response; Curve B shows response of subsequent runs. 25°C data.

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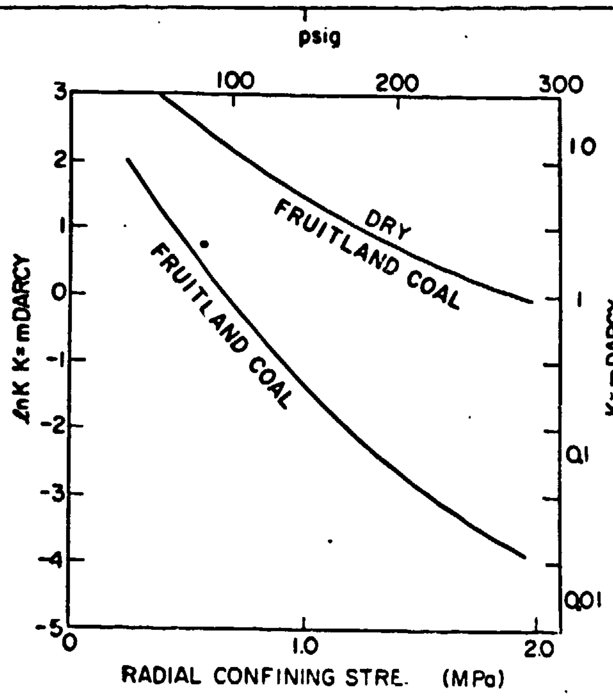

Figure 3: Effects of moisture content on the variation of CO_2 permeability with confining stress. Upper curve following moisture removal. Sample: Fruitland Seam, Sage Pit (Western Coal Company)

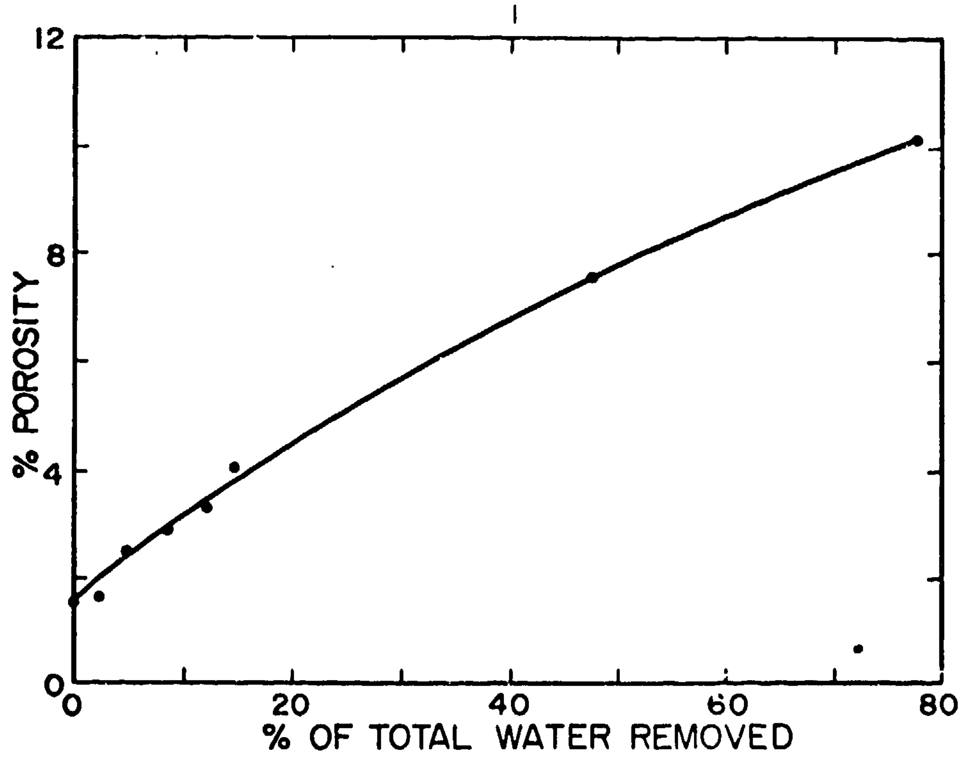

Figure 4: Generation of porosity in subbituminous coal (Fruitland) as a result of moisture removal. Boyle's Law porosimetry measurements made on coal cylinder with two open faces. Moisture removed by heating at 90°C. All measurements made at 25°C. Measurement gas, CO_2

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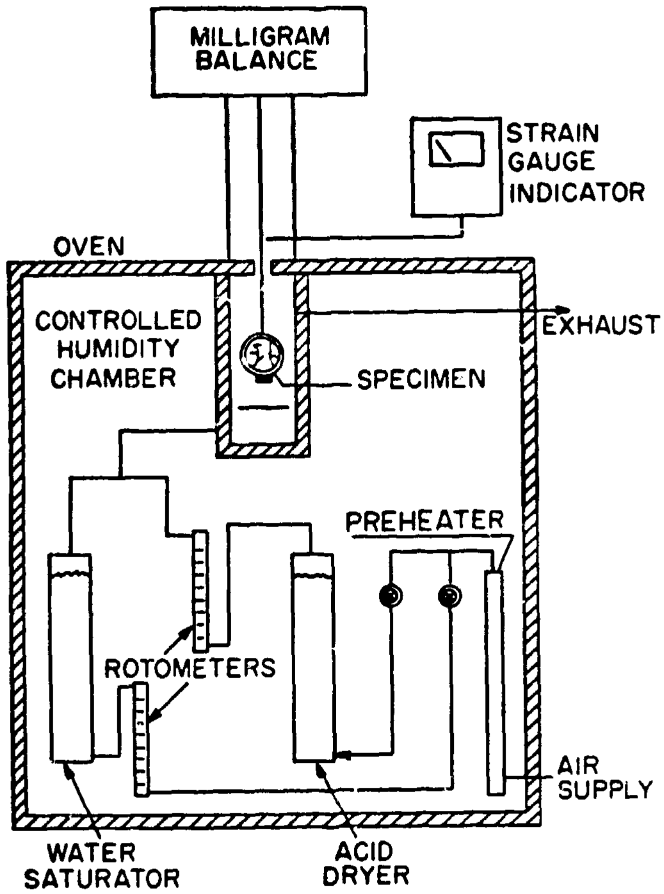

Figure 5: Schematic of apparatus to measure adsorbed water removal rates

Sample, right cylindrical section, is stressed. Simultaneous measurements of stress and weight are made as gas at controlled temperature and relative humidity is passed through measurement chamber. Gas mixture is blended from two streams - saturated through water scrubber and dehydrated through concentrated H_2SO_4. Varying flow proportions permit gas with compositions with 0 - 100% RH.

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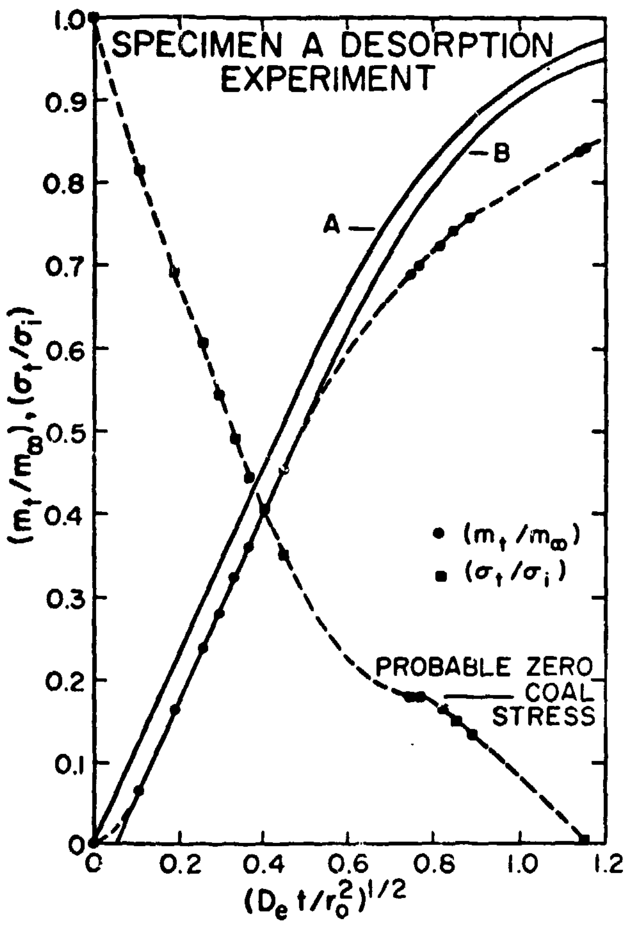

Figure 6: Kinetics of moisture removal. M_t= loss of weight at any time, t. M_∞ = total mass loss when sample is completely dry; σ_i = initial stress level. D_e = effective diffusion coefficient; r_o = half sample thickness; t = time. Real time elapsed is about 8 hr.

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the coal structure by irreversible changes in the coal structure that occur during drying. It is important to also consider the influence of stress on the removal rates of adsorbed moisture.

We assume here that the sole driving force during adsorbed water removal is a concentration gradient. Consequently we are describing the movement of water through the pore structure of coal to some larger flow feature, i.e., to the place where "adsorbed" water is transferred to "excess" water. This process was studied by measuring the weight of coal sections maintained under compressive stress in a constant humidity environment.

The method used is shown in Figure 5. A slab of coal is inserted into a cylindrical clamp and suspended from a balance. Simultaneously, measurement of the drying rate (mass) and of the compressive stress (strain) level were made. Measurements are made while the sample is suspended at constant temperature and constant humidity. The perimeter of the slab is covered with Teflon tape which both seals the surfaces and distributes the compressive stress throughout the specimen. By sealing the outer edge, mass transfer is essentially one-dimensional, and in a direction perpendicular to the plane of stress. Full descriptions and details of experimental procedure have appeared (10).

Table 1 gives a summary of results that were determined with these samples. In each case, moisture removal resulted in destressing the sample. Consequently, the stressed, initial results predict the rate of adsorbed moisture removal. Typical data are shown in Figure 6.

Figure 6 shows that initially the dimensionless drying curve exhibits a linear portion. Data also show that the stress level is rapidly reduced and approaches zero stress by the time the drying is 60% completed. (This experiment simulates the situation where lithostatic stress does not continuously follow the coal sample but, at some void value, the overburden becomes selfsupporting). Curve A, plotted as M_t/M (see Figure 5) versus dimensionless time shows system response to a step change in humidity. Curve B is another theoretical curve shifted ahead in time so that this result coincides with drying data obtained during the initial drying step. This time lag results from the fact that instantaneous change in surface concentrations do not occur due to the finite sample size. The experimental data agrees well with theory during the initial part of the experiment. However, after a dimensionless time of about 0.5, the data deviates sharply from predicted values, slowing markedly from predictions generated with a constant diffusion coefficient. This deviation from theory suggests that the last part of moisture is removed from disconnected pockets and removal involves other, more energetic kinetic processes than drying. Crank (11) noted that this test procedure and analysis yields mean diffusion coefficients over a range of saturation that yields a linear portion of the drying curve. Consequently, these data (Table I) are evaluated for this initial region that describes the stressed removal rates for adsorbed water.

Conclusions

Because of the altered flow paths which are generated concurrently with moisture removal, drying is a key step for controlling mass

TABLE I

Effect of Stress on Adsorbed Moisture Removal

Sample	Half Thickness r_o	Temperature	Effective Diffusion Coefficient D_e	Initial Moisture Content	Mean Saturation	Initial Stress Level
	cm	°C	cm^2/sec	Wt %	C/C_o	MP_a
A	0.47	58	1.5×10^{-6}	7.4	0.78	3.4
C	0.43	98	4.2	7.8	0.90	3.9
C	0.43	96	6.4	8.6	0.77	4.5
C	0.43	68	0.88	9.1	0.84	1.4
C	0.43	98	4.0	8.6	0.92	0.12

Samples: State of Washington subbituminous coal. As received moisture, 10%; ash, 15%; fixed carbon, 41%: volatile matter, 36%.

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transfer in underground coal conversion. Mass transfer through the coal bed is thus significantly influenced by the moisture removal step, and the mass transfer kinetics, i.e., underground conversion. Water is held in two regimes, within the major flow channels (excess water) and within pores and other minor flow channels (adsorbed water). Dewatering involves drainage from both types.

The second stage of drying results in porosity generation. Depending upon the type of UCC process employed, this stage may be via forced convective transport of inert gases (heated CO_2, etc.) or product gases. The process of adsorbed moisture removal is identified as a series of steps, with the rate determining step dependent upon transport through the coal structure. Permeability increases with moisture loss and decreases with increasing compressive stress. The effective water diffusion coefficient decreases strongly with decreasing moisture concentration and decreasing stress.

These facts suggest that molecular events are the controlling step in adsorbed water removal from subbituminous coal. The laboratory data also suggest that compressive stress is significant in determining coal seam permeability i.e., in setting dimensions of the fissure network. Lithostatic stress is significant to the extent that it helps set diffusion path lengths, or characteristic fissure spacing. The effective diffusion coefficient is strongly affected by temperature changes, suggesting that water transport in coal is an activated process.

These results impact on optimum design for underground coal processing in several ways. They suggest the following:

1. Sharp boundaries exist between wet and dry coal. Due to the differences in permeability between wet and dry coal, efficient drying is possible in porous dry coal that will permit rapid exhaust of moisture from the interface.

2. Destressing is an important element in flow enhancement and drying. Once a section of coal is distressed by creating porosity, that void volume should distribute into surrounding coal sections opening additional fissures for viscous flow.

3. Diffusion rates are slow compared to convection. Mass transfer through coals that are stressed and saturated is inefficient. Transfer at significant rates requires a fissure network; drying requires a closely spaced network of connected viscous flow channels.

4. "Dewatering" using compressed gases removes little moisture. Dewatering requires stress release. Increasing pore pressure inflates the fracture system. Most likely, in virgin coal, this increased pressurization will simply redistribute fracture patterns through plastic deformation. Dewatering, should it occur, will simply be within the major fissure network. Due to the increased stress, adsorbed water loss may actually be retarded. Most water is retained.

5. Hot gas, convective drying offers best possibility to destress a coal seam for effective moisture removal. Shrinkage in subbituminous coal requires efficient removal of adsorbed water. Rates are accelerated with temperature. Once a region of high porosity has been generated, then lithostatic stress on coal surrounding that region can be dissipated. Continued heating should result in generation of a porous cavity, a necessity for underground processing. This suggests that underground seams should be best processed in a two-stage approach. (12, 13, 14). This process partially occurs in reverse combustion . The gasification that does result may well be set by the extent of distressing that occurs ahead of the gasification front.

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